Journal of Environmental Science, Health & Sustainability, 2025, 1(2), 131–143
https://journals.enviromindsolutions.com/jeshs
eISSN: 3067-946X

Article

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Elevated groundwater arsenic in deeper aquifers of the Brahmaputra Floodplains in Assam, India

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How to cite

Medhi, S., Choudhury, R., 2025. Elevated groundwater arsenic in deeper aquifers of the Brahmaputra floodplains in Assam, India. Journal of Environmental Science, Health & Sustainability, 1(2), 131–143.
https://doi.org/10.63697/jeshs.2025.10038

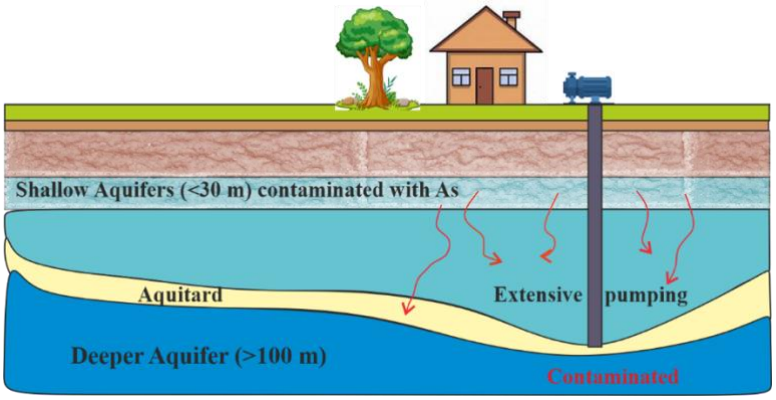
Article info

Received: 22 June 2025
Revised: 2 August 2025
Accepted: 10 August 2025
Published: 30 August 2025

Keywords

Arsenic
Groundwater
Deep aquifers
Brahmaputra floodplains
Irrigation pumping

Graphical abstract



Highlights

- *Elevated arsenic concentrations were observed adjacent to Naga foothills.*
- *Deeper aquifers are contaminated with arsenic concentrations >10 µg/l.*
- *Co-occurrence of elevated As, Fe and Mn concentrations in the deeper aquifer.*
- *The contamination of deeper aquifers may be attributed to groundwater pumping.*

Abstract

The deeper aquifers are considered safe for human consumption in various parts of South Asian countries, including the Ganges floodplains of India, and Bangladesh. The present study explored the distribution and controls of groundwater arsenic (As) in deeper aquifers (depth >80 m) as well as shallow aquifers (depth <30 m) that are considered a major source for drinking and irrigation purposes in Brahmaputra floodplains, India. We collected groundwater samples from the two most As-contaminated districts in Assam, India. About 87% of sampled wells in deeper aquifers had As concentration above the WHO permissible limit of 10 µg/l, with a mean concentration of 61 µg/l and 82 µg/l during pre- and post-monsoon seasons, respectively. In addition, about 55% of sampled wells in shallow aquifers had As concentration >10 µg/l, with a mean concentration of 47 µg/l and 56 µg/l during pre- and post-monsoon seasons, respectively. Various physico-chemical parameters, viz; pH, EC, TDS, Ca^{2+}, Na^{+}, K^{+}, Mg^{2+}, Cl^{-}, HCO_3^{-}, NO_3^{-}, and SO_4^{2-} were analyzed to elucidate the groundwater hydrochemistry of the region. The Piper plot indicated Ca-HCO_3 and Na-HCO_3 as the major water types in the study area in both deeper and shallow aquifers. The Gibbs plot depicted the rock-water interaction as a major source that governs the groundwater chemistry in the region. The regional scale study indicated that the vertical movement of elevated As from shallow aquifers to deeper depths, coupled with extensive irrigation pumping, could be a probable cause of As in the deeper aquifers.

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Handling Editor: Dr. Prosun Bhattacharya with assistance from Dr.Alper Baba

1 Introduction

Groundwater has been considered as the most pristine form of drinking water for ages, and access to fresh water is the elemental human right and is a part of constructive policy for safeguarding human health (Mukherjee et al., 2021; Nath et al., 2025). However, groundwater can be naturally and/or anthropogenically contaminated with trace elements like arsenic (As), iron (Fe) and manganese (Mn) (Mukherjee et al., 2008; Mahanta et al., 2015; Santha et al., 2022). Nonselective use of surface water bodies, such as rivers, lakes, and ponds, for discharging untreated industrial effluents coupled with geogenic contamination has placed a lot of people at acute health risk (Mukherjee et al., 2011). Millions of people globally, especially the population of South Asian countries, including India, are affected by prolonged intake of As-contaminated groundwater (>10 µg/l, above WHO permissible limit) (Bhattacharya et al., 1997; Mahanta et al., 2015; Choudhury et al., 2017; Shaji et al., 2020; Nath et al., 2022; Medhi and Choudhury, 2024).

Arsenic has been considered as one of the most harmful elements that has high carcinogenic potential to create severe impact on human health (ATSDR, 2019). In India, various regional scale studies have reported As-contaminated groundwater, for examples, in the states of Assam (Singh, 2004; Mahanta et al., 2015; Choudhury et al., 2017; Verma et al., 2019; Nath et al., 2022; Choudhury et al., 2024), Bihar (Chakraborti et al., 2003; Ivy, 2025), West Bengal (Bhattacharya et al., 1997; Nickson et al., 2007; Mukherjee et al., 2008; Sarkar et al., 2022; Mukherjee et al., 2024), Uttar Pradesh (Ahamed et al., 2006; Kumar and Pati, 2022; Soni et al., 2024), and Punjab (Kumar et al., 2019). The alluvial sedimentary deposits, particularly in Ganges-Brahmaputra floodplains, are identified as being the worst affected regions with elevated As in groundwater (Mukherjee et al., 2021). Consequently, searching for a safe and sustainable source of drinking water remains the biggest challenge in these areas.

In Brahmaputra floodplains, elevated As are thought to be usually found in shallow aquifers (Aind et al., 2025). Several studies (Burgess et al., 2010; Khan et al., 2019; Biswas et al., 2012) particularly in West Bengal, India indicated that deeper aquifers, mostly greater than 100 m in depth, are comparatively safe from elevated As concentration. Additionally, a study in Bangladesh, have reported that deeper aquifers are safer in terms of As contamination and can be used as an alternate source of drinking water (JICA, 2002; von Bromssen et al., 2007). The studies reported that the deeper aquifers of Bangladesh with low As concentration, comprised of Pleistocene sediments that were oxidized and were brownish in color, and are overlayed by aquifers of Holocene sediments that was enriched with As and was more reduced and darker grey in color (von Bromssen et al., 2007; Mukherjee et al., 2011). A study in Lampang Basin, Northern Thailand, reported higher As concentrations in deeper aquifers (Bashkin and Wongyai, 2002; Santha et al., 2022). However, similar hydrogeochemical conditions may not be present in the deeper aquifers in the Brahmaputra Basin. The term deeper aquifers in this study area are applied to those public water supply submersible wells, that pump the groundwater from the deeper depth, which usually varies from 80 to 200 m.

Therefore, the present study aimed to investigate the distribution of As in both deeper (>80 m below ground level, bgl) and shallow aquifers (<30 m bgl) during pre- and post-monsoon seasons and to find the probable cause of elevated As in the deeper aquifers of the study area. This study will provide insights into seasonal variations in As contamination as well as its vertical variations. The outcome of this study will likely contribute to the development of sustainable groundwater extraction strategies. Furthermore, the study will help identify potential risks to communities relying on deeper aquifers for drinking water.

2 Study area

2.1 Geography

The study area encompasses two districts of Assam (Jorhat and Golaghat) in the southern bank of Brahmaputra floodplains covering a total area of 6,352 km^2 in between the graticules of 26°45'28.34" N – 26°3'26.67" N latitude and 94°12'35.36" N – 93°57'47.47" E longitude (**Fig. 1**). Out of 7,702 habitations in rural areas, 4,976 habitations are receiving water from the Public Water Supply Scheme (PWSS) wells as of 2023 (PHED, 2024). The PWSS wells extracts water from the deeper aquifer and supply treated water for drinking as well as for other domestic purposes.

2.2 Geology

Assam is confined by the Himalayan Mountain belt in the north, Mishmi hills of Arunachal Pradesh in the east, and the Naga hills of Assam-Arakan fold and thrust belt, also known as the belt of Schuppen in the south. The Brahmaputra River flows from northeast to southwest in the middle of the state (Gogoi et al., 2022). Various tributaries of Brahmaputra River that have drained the Himalayas and Mishmi hills (Subansiri, Jia Bharali, Dikrong, Dibang, Dihang, and Puthimari) in the north and Naga hills and Meghalaya (Dhansiri, Dikhow, Dihing, Jhanji, Kopili, Buridihing, Dibru, and Sonai) in the south have created a large network of channels in the downstream and finally merge with the Brahmaputra (Gogoi et al., 2022). The study area, Jorhat and Golaghat districts, is covered with alluvial sediment of Quaternary age (Baviskar et al., 2015; Choudhury et al., 2018).

2.3 Hydrogeology

The average groundwater level in the study area is under 5 m bgl, with the water level varying from 0.41 to 3.07 m bgl during the pre-monsoon season and 0.56 to 3.4 m bgl during the post-monsoon season (CGWB, 2013). In the study area, shallow groundwater occurs in unconfined to semi-confined conditions, while in deeper depth, groundwater occurs in

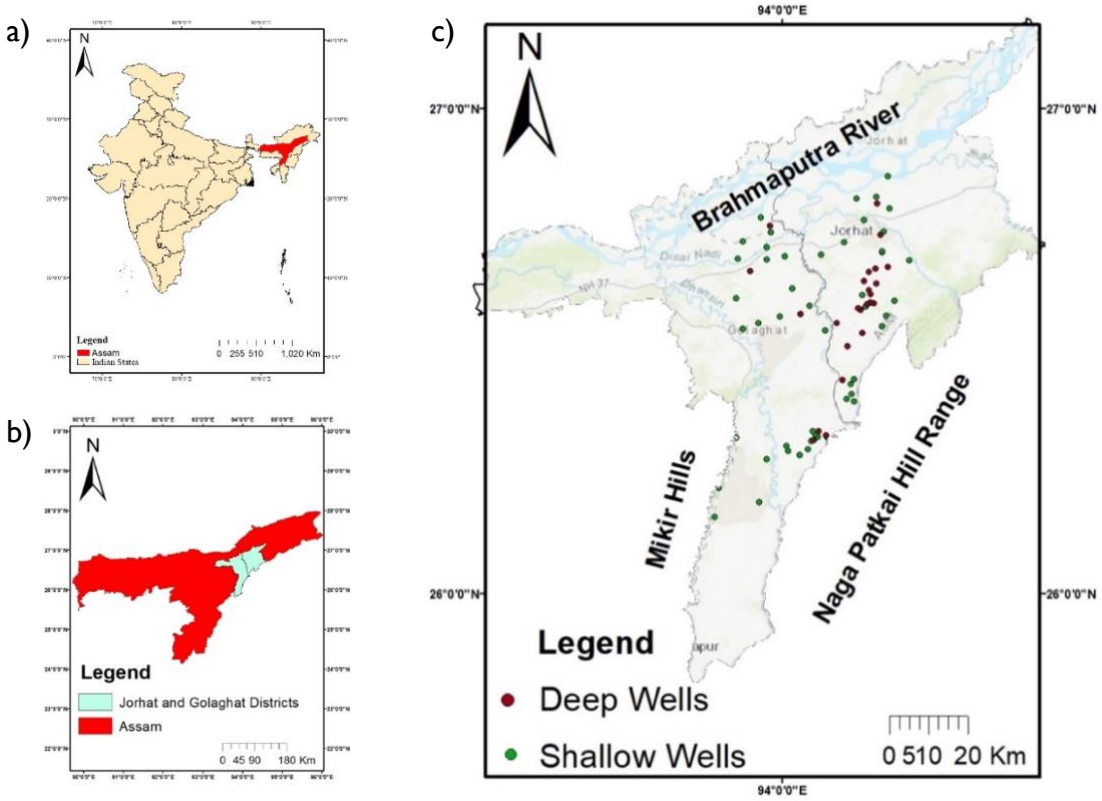

Figure 1. *Map showing: (a) India, (b) the state of Assam, and (c) the study area with locations of groundwater samples collected from shallow and deep wells.*

semi-confined to confined conditions (CGWB, 2013). Subsurface data obtained up to a depth of 300 m bgl specified the presence of three to four copious systems (Choudhury et al., 2020). In the southern part of the study area, the thickness of the aquifers is reduced due to the decrease of coarse to medium sand particles and an increase of finer grains like clay and silt. Near the Brahmaputra River, up to a depth of 400 m, five to six aquifer systems can be seen of varying thicknesses (CGWB, 2013). The hydraulic conductivity of the aquifers in the study area ranged from 10 to 60 m/day while its transmissivity value ranges from 415 to 5,632 m²/day (CGWB, 2013; CGWB, 2022).

3 Materials and methods

3.1 Sampling strategy

Twenty-four PWSS wells were selected from the study area to monitor As concentrations in the deeper aquifers (>80 m). In 2022, unfiltered groundwater samples were collected to measure dissolved As concentration during both pre- and post-monsoon seasons. Along with the PWSS wells, 50 groundwater samples from shallow aquifers (<30 m, depth ranging widely from 5 m to 30 m) covering the two districts (Jorhat and Golaghat) were also collected after purging the wells for 5 min to get the representative groundwater samples. The groundwater samples were acidified with 6N HNO_3 and were collected in 20 ml HDPE bottles and were stored at 4°C before analyzing the samples in the laboratory.

3.2 Chemical analysis

Eutech (Cyber scan series 600) portable meter was used to determine the physical parameters, viz; pH, EC, and TDS on-site. Cations and trace elements (As, Fe, and Mn) were analyzed using inductively coupled plasma optical emission spectrometry (ICP-OES, Thermo scientific *i*CAP PRO) in the Hydrogeology Laboratory of IIT Kharagpur. The precision of the instrument was observed to be ~3%. The quality check and the calibration of the instrument were done using the comparable standard of NIST. For alkalinity, the groundwater samples were titrated against 0.1N HCl. Chloride was analyzed using argentometric method following the procedure of APHA (2017). UV-VIS spectrophotometer (UV-1900i) was used to determine SO_4^{2-} and NO_3^- at wavelengths 420 nm and 220 nm, respectively. Saturation index (SI) was calculated using PHREEQC software.

4 Results and discussions

4.1 Hydrochemistry

The pH values for both shallow and deeper wells indicate that the groundwater is neutral to alkaline in nature, with similar average pH values of 7.5 and 7.4, respectively. The

concentration of EC and TDS is nearly 30% higher in deeper wells than in shallower wells. Additionally, the concentration of TDS and EC were seen higher during pre-monsoon season in both deeper and shallower aquifer. This may be attributed to the effects of evaporation, which can lead to the elevated concentration and subsequent enrichment of dissolved solutes in the groundwater system (Manikandan et al., 2020).

The major ion concentration was found to be slightly higher in the deeper wells than the shallower wells (**Table 1**). This could be due to considerably higher residence time with the subsurface material with depth (Fenta et al., 2020). The major ion concentration seems to vary with depth in the studied region. The concentration of major cations and anions of shallow aquifer are in the order $Ca^{2+} > Na^{+} > Mg^{2+} > K^{+}$ and $HCO_3^{-} > Cl^{-} > SO_4^{2-} > NO_3^{-}$, while for deeper aquifers the concentration of major cations and anions are in the order $Na^{+} > Ca^{2+} > Mg^{2+} > K^{+}$ and $HCO_3^{-} > Cl^{-} > SO_4^{2-} > NO_3^{-}$. The higher concentration of Na^{+} compared to Ca^{2+} in deeper wells could be attributed to the cation exchange process, where Na^{+} is interchanged with Ca^{2+} and Mg^{2+} (Fenta et al., 2020). For deeper aquifers, the molar ratio of Na/Cl during pre-monsoon ranged from 0.18 to 4.97 with mean concentration of 1.59. While during post-monsoon the molar ratio of Na/Cl ranged from 0.04 to 5.26 with mean concentration of 1.47. For shallow aquifers, the mean Na/Cl ratio is greater than 1 during pre-monsoon (range: 0.05 to 3.65, mean: 1.1) and post-monsoon seasons (range: 0.01 to 8.9, mean: 1.3). The Na/Cl molar ratio >1 mostly results in silicate weathering/cation exchange, whereas <1 mostly results in reverse ion exchange process (Manikandan et al., 2020). In the present study, the Na/Cl molar ratio >1 indicating dominance of cation exchange and silicate weathering processes. However, some samples fell below the equiline indicating the influence of reverse ion and carbonate dissolution during both the seasons (**Fig. S1**). Similar results were shown in the bivariate plot of Na^{+} and Ca^{2+} for both deeper and shallow aquifers (**Fig. S2**). Moreover, the weathering of Na^{+} bearing silicate minerals may contribute to elevated Na^{+} concentrations in the deeper aquifers (Fenta et al., 2020; Medhi and Choudhury, 2024).

Hydrochemical facies were determined to understand the water type of the study area. The Piper plot indicated that the groundwater of the study area is mostly Ca-HCO_3 type and Na-HCO_3 type in both deeper and shallow wells (**Fig. 2**). As depicted in the Gibbs plot, rock-water interaction seems to be the dominant process that controls the overall chemistry of groundwater in both shallow and deeper wells (**Fig. 3**). The bivariate plots of Ca^{2+}/Na^{+} vs HCO_3^{-}/Na^{+} and Ca^{2+}/Na^{+} vs Mg^{2+}/Na^{+} indicate silicate weathering is predominantly responsible to control the chemistry of the groundwater in both deeper and shallow aquifers during both the pre- and post-monsoon seasons, together with some influence of carbonate dissolution processes (**Fig. 4**). The concentration of NO_3^{-} and SO_4^{2-} in the rainwater is naturally low, however there might be substantial anthropogenic impact from septic tanks. Additionally, there might be possible reactions of

Table 1. *Descriptive statistics of water quality parameters in the study area, and bdl stands for below detection limit.*

Parameter	Deep wells						Shallow wells					
	Pre-monsoon			Post-monsoon			Pre-monsoon			Post-monsoon		
	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean	Max	Min	Mean
pH	8.0	5.9	7.2	8.1	7.3	7.7	8.1	6.3	7.3	8.2	7.1	7.6
EC (µs/cm)	1,255	259	643	850	110	421	1,019	39	425	659	99	321
TDS (mg/l)	803	224	455	426	57	211	510	19	218	328	49	161
Ca^{2+} (mg/l)	64	9.6	25	99	22	50	77	0.41	21	151	26	56
Na^{+} (mg/l)	151	11	50	209	1.8	50	97	1.7	30	132	0.50	27
Mg^{2+} (mg/l)	16	1.2	8.5	38	0.02	19	11	0.73	2.3	33	1.2	14
K^{+} (mg/l)	13	1.3	4.6	8.5	0.09	1.5	18	2.0	8.9	14	0.09	1.8
HCO_3^{-} (mg/l)	517	133	318	300	100	217	467	100	233	400	100	201
Cl^{-} (mg/l)	220	23	43	51	23	35	131	34	47	131	17	38
NO_3^{-} (mg/l)	7.0	0.65	3.5	3.3	0.1	1.2	2.4	0.49	1.1	2.0	bdl	0.46
SO_4^{2-} (mg/l)	44	bdl	3.6	52	bdl	4.3	13	bdl	1.5	50	bdl	5.6
As (µg/l)	134	bdl	61	169	2.1	83	480	0.07	47	0.64	531	56
Fe (mg/l)	25	0.11	5.6	37	0.09	7.6	41	0.08	11	39	0.11	12
Mn (mg/l)	2.6	0.02	0.35	3.4	0.002	0.39	9.1	0.002	0.66	8.2	0.007	0.79

anthropogenic organic matter with nitrate and ferric oxyhydroxides after nitrate reduction resulting in lower concentration of NO_3^- and SO_4^{2-} in the groundwater of the present study area (McArthur et al., 2012).

4.2 Groundwater flow direction

The groundwater flow direction was established using the water table data that was delineated from the data collected in the field from deeper wells during pre-monsoon season (**Fig. 5**). The groundwater flow direction is controlled by the topography of the region. The flow direction was from higher terrain in the south and southwest towards the lower terrain in the north and northeast (Choudhury et al., 2018). Apparently, the higher concentration of As is seen in the southern part of the study area, adjacent to the Naga foothills, and a lower concentration of As near the Brahmaputra River towards the north, potentially indicating the source of As-bearing sediments are being transported from the Naga foothills and accumulated in the surrounding aquifers. The groundwater flow further implies that the As-enriched groundwater might be transported to other areas and slowly contaminating the areas that are currently low in As concentrations.

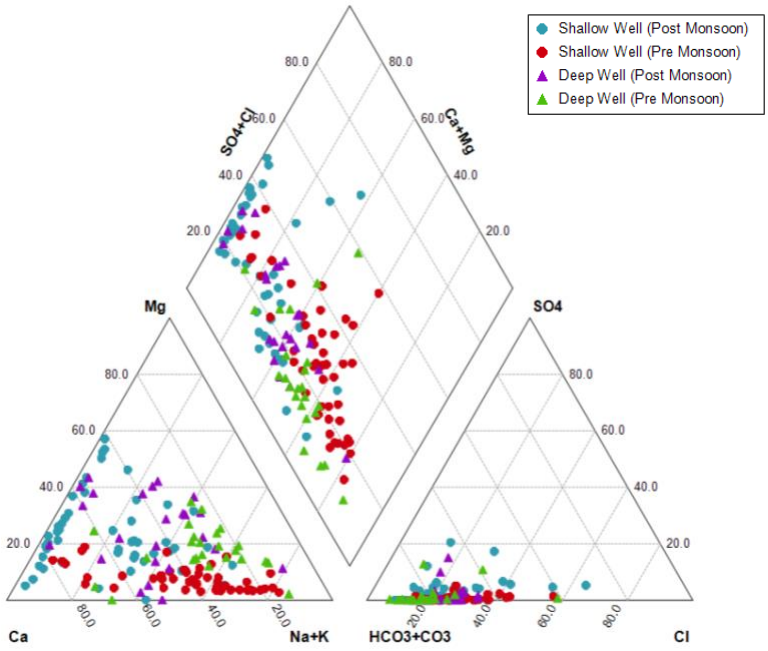

Figure 2. *Piper plot showing the chemical composition of groundwater samples and the water types of the study area.*

4.3 Distribution and source of arsenic

The spatial distribution of As in deeper and shallow aquifers in both pre- and post-monsoon seasons indicate that the aquifers in the southern part of the study area, adjacent to the Naga foothills, are highly contaminated with As (**Figs. 6 & 7**). Presence of elevated As in deeper aquifers was observed in most of the wells where the concentration ranged from bdl (below detection limit) to 134 µg/l, with a mean concentration of 61 µg/l during the pre-monsoon season, while during the post-monsoon, it ranged from 2.1 µg/l to 169 µg/l, with a mean value of 82 µg/l. Around 87% and 84% of analyzed samples showed As concentration beyond the WHO permissible limit of 10 µg/l during the pre- and post-monsoon seasons, respectively. While around 66% of deep aquifers showed As concentration >50 µg/l, making them unfit for human consumption. On the other hand, 55% and 62% of shallow aquifers had an average concentration of 47 µg/l (range: 0.07 to 480 µg/l) and 56 µg/l (range: 0.64 to 531 µg/l) during pre- and post-monsoon seasons, respectively.

Verma et al. (2019) investigated the sedimentological and geochemical characteristics of aquifers in the southern

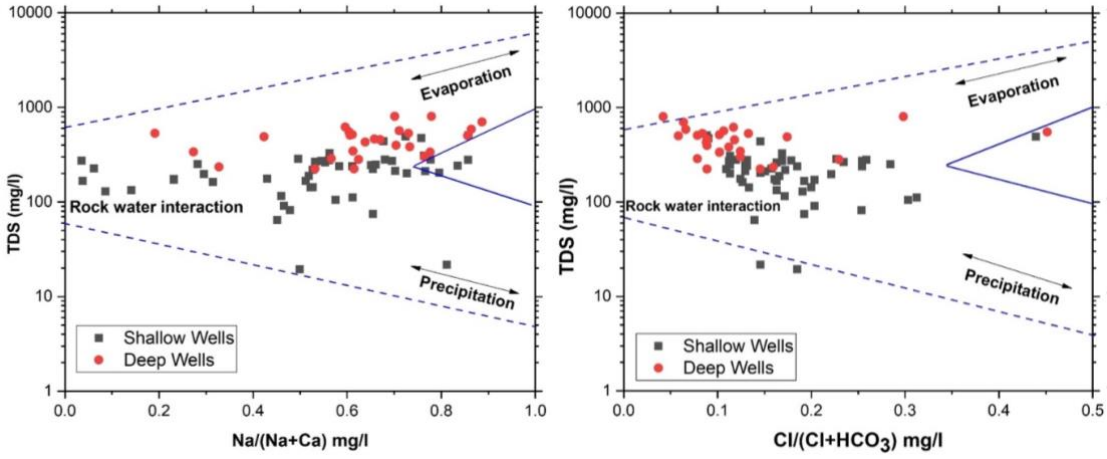

Figure 3. *Gibb's plot of shallow and deep groundwater samples in the study area.*

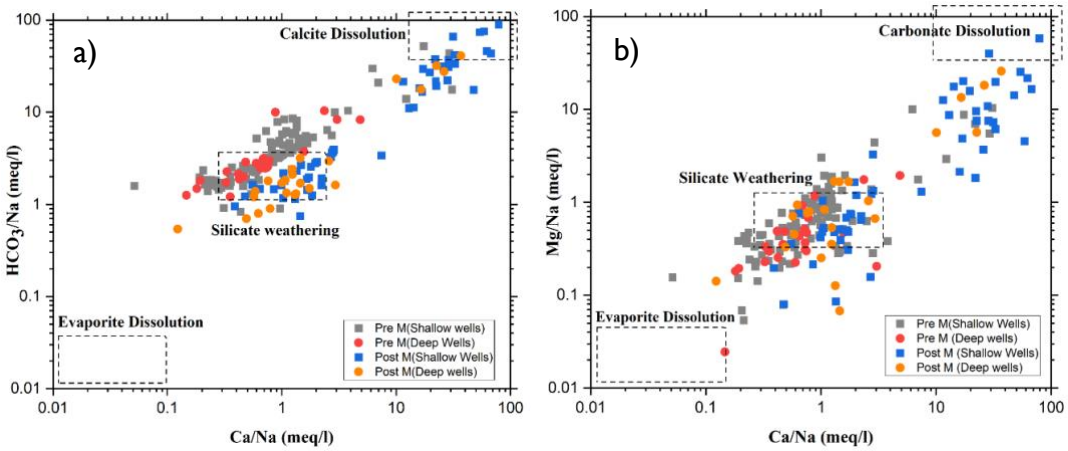

Figure 4. *Bivariate plots of: (a) Ca^{2+}/Na^{+} vs HCO_3^{-}/Na^{+}, and (b) Ca^{2+}/Na^{+} vs Mg^{2+}/Na^{+} in shallow and deep wells during pre- and post-monsoon seasons.*

part of the Brahmaputra floodplains and identified a strong tectono-morphic influence. Their analysis revealed the dominance of Fe/Mg-rich aluminosilicates, along with heavy minerals and clays such as titanite and ilmenite. These minerals are likely the result of prolonged weathering of mafic and ultramafic source rocks, particularly from the Naga ophiolitic belt. Their study proposed that intense chemical weathering of As-enriched mafic minerals such as olivine, chamosite, and titanite combined with prevailing reducing conditions in the aquifer facilitated the mobilization of As into groundwater. Mahanta et al. (2015) and Choudhury et al. (2018) reported the presence of thick clay layers in aquifers near the Naga foothills. These clay layers likely function as As sinks, accumulating As over time. However, under favorable redox conditions, As is gradually released into the surrounding groundwater. In contrast, aquifers situated closer to the Brahmaputra River are primarily composed of fine to medium-grained sand, which enhances aquifer permeability and supports rapid groundwater recharge and flushing. Choudhury et al. (2018) observed that improved flushing capacity contributes to lower As levels in the aquifers adjacent to the present-day river channel.

4.4 Release mechanism of arsenic

Previous studies in Ganges and Brahmaputra basins suggested reductive dissolution of iron oxy hydroxide (FeOOH) as the primary mechanism of As release into the groundwater (Bhattacharya et al., 1997; McArthur et al., 2001; Dowling et al., 2002; Mukherjee et al., 2008; Mahanta et al., 2015). However, other authors have suggested that oxidation of As-rich pyrite can also lead to As release into the aquifers (Das et al., 1996). Mild oxic conditions can also favor reductive dissolution of As (van Geen et al., 2004). However, the release processes of As can be complicated, if there are complex patterns of oxidation, reduction, and reoxidation of Fe and S species (Zheng et al., 2004). In addition, As that is released into the groundwater through reductive dissolution can be absorbed again by the partly reduced FeOOH minerals (McArthur et al., 2004).

Globally, several authors have reported a strong correlation between As and Fe, which confirms the reductive dissolution of FeOOH as the primary As release mechanism. However, the present study showed a weak correlation between As and Fe, with $r^2 = 0.0003$ for shallow aquifers and $r^2 = 0.05$ for deeper aquifers (**Figs. S3a, S4a**). Mahanta et al. (2015) also found a similar result in the southern bank of Brahmaputra River, with r^2 value of 0.2. The weak association between As and Fe observed in this study may be due to reactive behavior of Fe minerals, as it tends to precipitate under reducing conditions rather than remaining dissolved in groundwater (Medhi et al., 2024). This

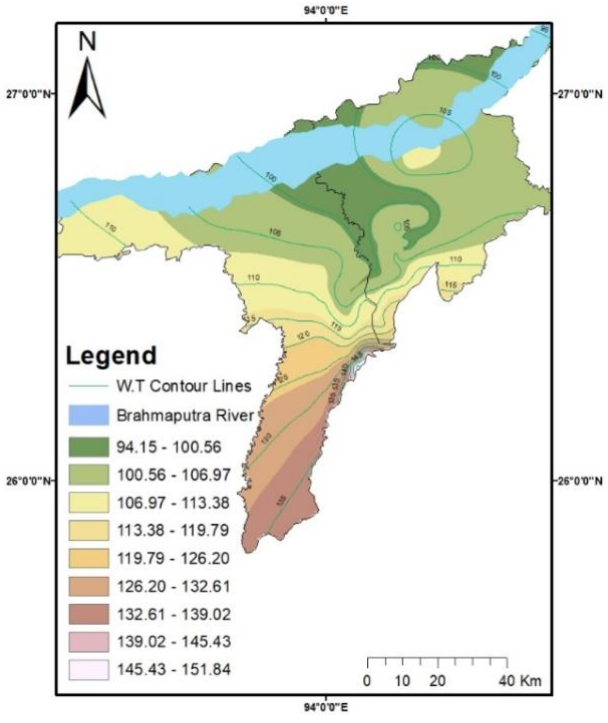

Figure 5. *Regional-scale groundwater flow direction in the study area.*

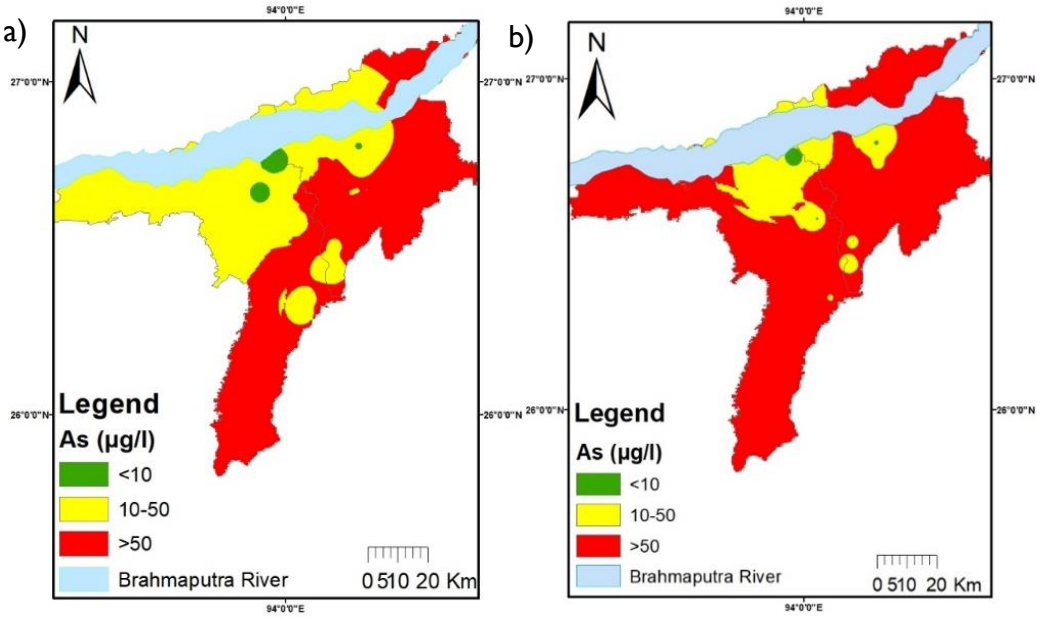

Figure 6. *Arsenic distribution in deeper aquifers during: (a) pre-monsoon, and (b) post-monsoon seasons.*

process often results in the formation of minerals such as siderite ($FeCO_3$), pyrite (FeS_2), and vivianite ($Fe_3(PO_4)_2 \cdot 8H_2O$), as previously reported by several authors (Mahanta et al., 2015; Choudhury et al., 2017; Mukherjee et al., 2008; Guo et al., 2013; Sracek et al., 2018). Moreover, the saturation indices (SI) of siderite and rhodochrosite calculated for the present groundwater samples were found to be 2.7 and 1.5, respectively. These values indicate that both minerals are supersaturated (SI > 1), suggesting favorable geochemical conditions for their precipitation. The precipitation of siderite and rhodochrosite may have contributed to the observed de-coupling of Fe, As, and Mn in the groundwater system (Sracek et al., 2018).

Correlation of As with various other parameters, viz, Mn (r^2 = 0.006, shallow wells and r^2 = 0.07, deeper wells) (**Figs. S3b, S4b**), SO_4^{2-} (r^2 = 0.0005, shallow wells and r^2 =

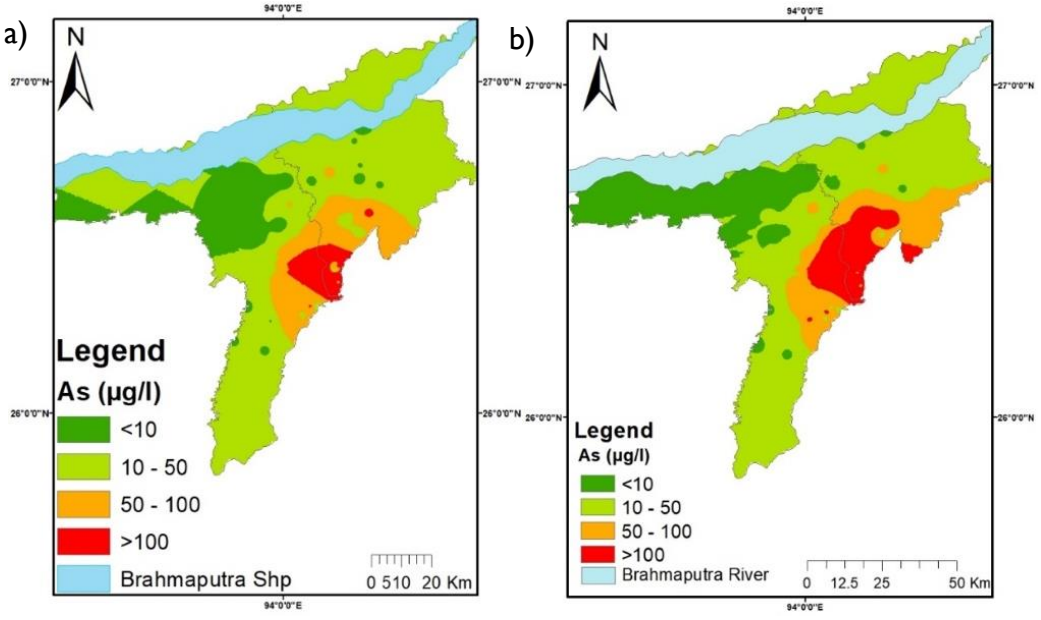

Figure 7. *Arsenic distribution in shallow aquifers during: (a) pre-monsoon, and (b) post-monsoon seasons.*

0.0014, deeper wells) (**Figs. S3c, S4c**), NO_3^- (r^2 = 0.008, shallow wells and r^2 = 0.25, deeper wells) (**Figs. S3d, S4d**), and HCO_3^- (r^2 = 0.0014, shallow wells and r^2 = 0.081, deeper wells) (**Figs. S3e, S4e**) did not show any notable relationships indicating that there might be multiple geochemical processes taking place in releasing As into the groundwater (Mahanta et al., 2015; Choudhury et al., 2018). Release of As into the deeper aquifers of Brahmaputra floodplains through pH-dependent sorption process is unlikely, as poor correlation was also observed between As and pH (r^2 = 0.06, shallow wells and r^2 = 0.0001 deeper wells) (**Figs. S3f, S4f**).

4.5 Depth profile of As, Fe, Mn and SO_4^{2-}

Variation in the concentration of groundwater As, Fe, Mn, and SO_4^{2-} with depths is illustrated in **Figure 8**. No specific depth correlation is seen between As with Fe, Mn, and SO_4^{2-}, as high As (>10 µg/l) did not essentially coincide with high Fe and Mn concentration and low SO_4^{2-} concentration. Elevated groundwater As concentration (>50 µg/l) is mainly found to be intense from shallow sub-surface to 150 m in depth. Intricate precipitation-dissolution processes coupled with decomposition of organic matter could possibly be the reason for differential concentration of these trace elements in the groundwater of the study area (Choudhury et al., 2017). The calculated saturation indices for siderite (SI = 2.7) and rhodochrosite (SI = 1.5) indicate that groundwater is supersaturated with respect to both minerals, suggesting thermodynamically favorable conditions for their precipitation. This supports the interpretation that elevated concentrations of Fe and Mn, in the presence of high HCO_3^-, may lead to the formation of Fe and Mn carbonates in the aquifer.

4.6 Groundwater pumping

A study in Vietnam reported that excessive extraction of groundwater from the As enriched groundwater in shallow aquifers to meet the requirement of the population demand

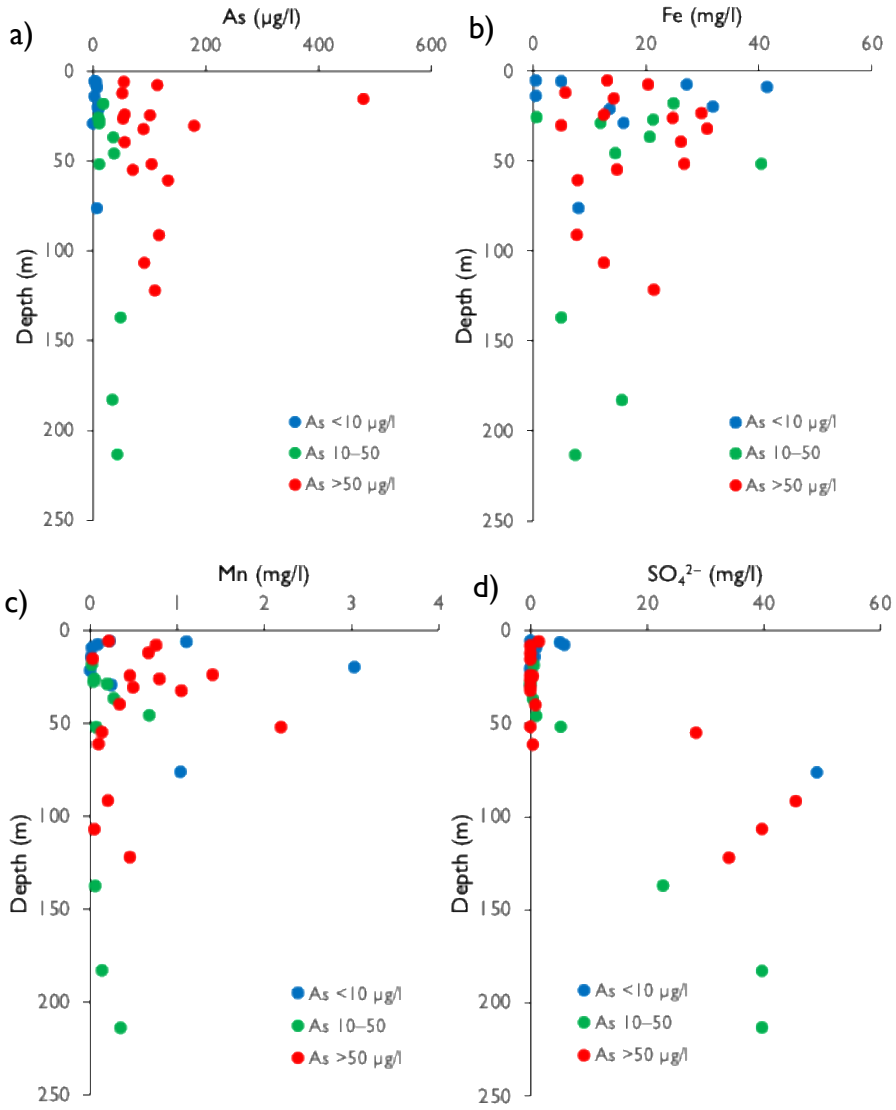

Figure 8. *Distribution of a) As, b) Fe, c) Mn, and d) SO_4^{2-} along the depth in the study area. Different colors signify the As*

has resulted in vertical transport of As, contaminating the deeper aquifers, which were initially free from elevated As concentration (Winkel et al., 2011). Other researchers have also mentioned the influence of irrigation pumping impacting the distribution of As in the deeper aquifers in the Ganges floodplains (Harvey et al., 2005; Mukherjee et al., 2007). In the present study area, 2,90,214 ha of land is covered under agricultural land, which includes various cropping practices. To meet the water demand for cultivation, deep irrigation wells with an average depth of 150 m are used to pump water to the agricultural field. These wells typically run for 5 to 6 hrs every day during Kharif or monsoon seasons, i.e., in the month of June and July, and 3 to 4 hrs every day during Ravi or post-monsoon seasons, i.e., in the month of October and November. According to the Irrigation Department's annual report (2022-2023), 20 HP pumps with a discharge rate of 160 l/sec are used to extract water from these irrigation wells (Irrigation Department, 2023). The groundwater As concentration of the PWSS (depth: 80 to 200 m) from the study area ranged from 2 to 169 µg/l with an average value of 82 µg/l, where 66% of the sampled PWSS had As concentration >50 µg/l, indicating the probable impact of irrigation pumping on the occurrence of elevated As in deeper aquifers (Mukherjee et al., 2011). Mukherjee et al. (2007) hypothesized that over the last few decades, human-induced modifications to groundwater flow patterns may have significantly raised recharge rates. This increased recharge can lead to increased downward transport of organic carbon and dissolved oxygen in areas lacking intermediate-depth aquitards, which may interfere with redox-sensitive biogeochemical processes below the surface (Harvey et al., 2002; Harvey et al., 2005; Mukherjee et al., 2008). Furthermore, agricultural fields get a large amount of As that is extracted and pumped to the surface (Roychowdhury et al., 2005); further infiltration may cause the As that is not absorbed by crops to return to the subsurface (Mukherjee et al., 2011). Nevertheless, 4% of sampled PWSS that have depth greater than 200 m, also showed As concentration >50 µg/l, indicating an equal possibility of As being transported naturally such as impact of recharge during monsoon into greater depth, instead of being contaminated by the shallower aquifers due to pumping.

5 Conclusion

This study concludes that high As concentration (>10 µg/l) in both deeper and shallow aquifers were found mostly close to the Naga foothills. The average concentration of major ions in deeper aquifers during both pre- and post-monsoon seasons were $Na^+ > Ca^{2+} > Mg^{2+} > K^+$ and $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^-$. However, the concentration of cations and anions in shallow aquifers during pre- and post-monsoon seasons were $Ca^{2+} > Na^+ > Mg^{2+} > K^+$ and $HCO_3^- > Cl^- > SO_4^{2-} > NO_3^-$, respectively. The piper plot indicated Ca-HCO_3 and Na-HCO_3 water type dominated both shallow and deeper aquifers, where rock-water interaction was seen to be the major process that controlled the groundwater chemistry. The bivariate plots indicated silicate weathering and ion exchange as the major processes that influence the groundwater chemistry with some contribution from carbonate mineral dissolution and reverse ion exchange processes. Poor correlation of As with Fe and other redox sensitive elements such as Mn and SO_4^{2-} and HCO_3^- indicate that there is no single mechanism that releases As into groundwater. Additionally, the precipitation of siderite and rhodochrosite may have contributed to the observed de-coupling of Fe, As, and Mn in the groundwater system. The study further concludes that the groundwater extracted from deeper aquifers through motorized submersible wells (PWSS wells) are not safe for drinking in the study area. In most of the study areas, there might be probable mixing of the As-enriched groundwater in shallow aquifers with the groundwater of deeper aquifers. The withdrawal of groundwater for irrigation from the deeper depths close to the PWSS wells over the last several decades might have triggered the downward movement of As-enriched shallow groundwater, together with natural contamination during monsoon recharge. Instead of relying on the deeper aquifers, switching over to treated surface water for drinking would be a better solution in the study area.

6 Acknowledgements

The authors sincerely acknowledge the Department of Geology and Geophysics, IIT Kharagpur, for providing essential laboratory facilities and instrumental support. The present study did not receive any financial support.

7 Data availability statement

Supplementary data is provided. Additional data can be made available at reasonable request from the corresponding author.

8 Author contributions

SM: data curation, formal analysis, investigation, methodology, software, visualization, and writing – original draft. RC: conceptualization, investigation, project administration, supervision, and writing – review & editing. All authors approved the final version of the manuscript.

9 Conflict of interest

The authors declare no known conflict of interest related to this study.

10 Ethical statements

This study does not involve human or animal subjects. Ethical approval was not required for this research.

11 Copyright statement

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